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Reply to Office Action of: 03/06/2007
Family Number/Atty. Docket No.: P2003J087 / JCW-0306

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REMARKS / ARGUMENTS

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In response to Examiner's Office Final Office Action dated March 6, 2007, rejecting all claims, applicants file this Request for Continued Examination under 37 CFR §1.114 for the present application and offer the enclosed claim amendments and responses to Examiner's rejections for allowance of the present claims.

The Examiner is respectfully requested to contact applicants' attorney after review of the current claim amendments and responses below, so that an interview may be scheduled with the Examiner prior to issuance of the next office action.

CLAIM OBJECTIONS

The Examiner has objected to the use of the expression "SiO₂/Al₂O₃ ratio" in line 8 of claim 1 stating that applicants should replace this expression with the term "SiO₂/Al₂O₃ molar ratio" in the claims.

Applicant's Position

Per Examiner's request, the term "SiO₂/Al₂O₃ ratio" as used in prior Claim 1 has been amended to "SiO₂/Al₂O₃ molar ratio" to clarify its intent. Support for this amendment has been provided to the Examiner in applicants' prior Office Action response and the associated affidavit, dated December 12, 2006.

Therefore, applicants respectfully request the Examiner to reconsider and withdraw this objection.

FIRST REJECTION UNDER 35 U.S.C. 103

The Examiner has maintained his prior rejection of Claims 1-3 and 7-9 under 35 U.S.C. 103(a) as being unpatentable over Olson (6,488,741).

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Examiner's Position

It is the Examiner's position that Olson discloses a process of selectively adsorbing propylene in a mixture of propylene/propane through the use of zeolites having structures of a maximum of 8-member tetrahedral rings controlling the diffusion rate. Zeolites are those of the CHA (e.g., SSZ-13) and ITE structures.

It is Examiner's position that although Olson does not disclose a zeolite with a silica to alumina molar ratio of greater than 50 but less than 200, that it would have been obvious for one to use a zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio less than 200 (e.g., 199).

Applicant's Position

Applicants have presently amended Claim 1 to require the zeolite used as an adsorbent in the process to be a dealuminated zeolite. Support for this amendment may be found in Claim 4 as originally submitted. Claim 1 has also been presently amended limiting the zeolite as utilized in the process to a " $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than about 80 and less than 180". Support for this amendment may be found in paragraph [0027] of page 9 of the specification as originally submitted. It is applicants' position that the claims 1-3 and 7-9 are not obvious in light of Olson.

Olson specifically limits the disclosure and claims of the patent to silica to alumina ratios of at least 200 (see Olson, column 2, line 60-61 and Claim 1). In fact, it is stated in Olson that the zeolite utilized in the process of Olson "must have a Si:Al ratio of at least about 200" (column 2, lines 48-49), therefore explicitly teaching away from the use of a zeolite with $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio less than about 200.

A problem that exists with low silica to alumina ratio zeolites is the fact that at low $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios (i.e., below 200), very adverse process affects are seen. This can be seen in Figure 1 of the present application where a CHA zeolite (here H-SSZ-13) at low $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios ($\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio = 20 in Figure 1) results in severe

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polymerization of the propylene resulting in almost no recovery (i.e., desorption) of the desired propylene product.

In fact, it can be seen in the examples of Olson that zeolites of only extremely high $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios were tested. In Examples 1-10 only pure silica zeolites ($\text{SiO}_2/\text{Al}_2\text{O}_3 = \text{infinity}$) were utilized in the adsorption tests. In the remaining Examples 11 and 12, an HZSM-5 zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 3,300 was utilized in the adsorption testing of the examples. At lower Si/Al ratios as presently claimed, deleterious polymerization effects are encountered with the zeolites of the prior art.

Olson therefore explicitly limits that the $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of the zeolites utilized in his process must be above about 200 and does not provide a solution to the problem of how to utilize a zeolite with a low $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio in such a process. It is desirable to use a zeolite with a lower $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio as these zeolites are generally cheaper and more readily available than the zeolites disclosed in Olson. It is not that Olson suggests that a zeolite of less than 200 could not be prepared, but instead the awareness of the fact of the highly deleterious effects of using a zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of less than about 200 in the separations process. What has been discovered in the current invention is a non-obvious method for making a zeolite adsorbent with low $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios while still maintaining excellent adsorption/desorption properties (high component diffusivity ratios) and utilizing this new adsorbent in a separations process as claimed. Again attention is drawn to the fact that Olson utilizes zeolites in his examples with $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 3,300 and above to illustrate the process benefits of his invention due to the inherent problems experienced with these low $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio zeolites.

Therefore, one of ordinary skill in the art would be explicitly taught away by Olson from the use of a zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than about 80 and less than 180 as presently claimed for a separations process as presently claimed.

It is can be seen from the present application that the problem of excessive polymerization has been observed at low $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios (as shown in Figure

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1 of the current application) and the problem has been solved in the presently claimed invention by modifying the 8-ring zeolite adsorbent by dealuminating the zeolite and ion-exchanging the zeolite with an alkali metal cation. The claimed separations process utilizing the resultant zeolitic adsorbent significantly minimizes the amount of product polymerization and significantly improves the recovery rate of propylene even at low $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios (see Figures 1, 2, 3 and 7 of the present application).

Claim 1 as presently amended requires the use of a dealuminated, alkali metal cation containing zeolite for which it has been discovered that propylene recovery can be significantly increased while allowing the use of a low $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio zeolite. The discovery of the use of ion-exchanged alkali metal cation containing dealuminated zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than 80 and less than 180 to significantly improve the recovery of the adsorbed compound as discovered in the presently claimed invention is not taught nor suggested by Olson. As explained above, Olson does not teach a dealuminated zeolite and explicitly teaches away from the $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios presently claimed.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of Claims 1, as well as Claims 2-3 and 7-9, which depend from Claim 1 as presently amended.

SECOND REJECTION UNDER 35 U.S.C. 103

The Examiner has maintained his prior rejection of claims 4-6 under 35 U.S.C. 103(a) as being unpatentable over Olson (6,488,741) as applied to Claim 1 above, and further in view of either Eberly et al. (3,591,488) or Wakita et al. (6,579,347).

Examiner's Position

It is the Examiner's position that Olson discloses a process of selectively adsorbing propylene in a mixture of propylene/propane through the use of zeolites having structures of a maximum of 8-member tetrahedral rings controlling the

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diffusion rate. Zeolites are those of the CHA (e.g., SSZ-13) and ITE structures, but that Olson does not specifically disclose a step for dealuminating the zeolite by using steam. It is Examiner's position that both Eberly and Wakita teach a step of dealuminating a catalyst by steam and therefore it would have been obvious to one of ordinary skill in the art to have modified the Olson process by dealuminating the zeolite by steam as taught by either Eberly or Wakita.

Applicant's Position

It is applicants' position that one of skill in the art with the teachings of Olson and knowledge of the Eberly and Wakita patents would not be led to the present invention. In the arguments above, the limitations of prior Claim 4 have been presently incorporated into Claim 1 and Claim 4 has been canceled. Therefore, in the arguments below, Examiner's prior rejection of Claims 4-6 will be addressed in the context of presently amended Claims 1, 5, and 6.

Firstly, Olson does not teach the use of a separations process utilizing an 8-membered ring metal cation containing zeolite adsorbent with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than 80 and less than 180 that maintains a diffusion rate for a first component at least 50 times greater than the diffusion rate for a second component as is presently claimed. Therefore, due to the problems inherent with the Olson zeolites at $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios less than about 200, one would not be motivated to utilize a zeolite adsorbent with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than 80 and less than 180 as presently claimed regardless of the processes by which the final zeolite is derived.

Secondly, as stated by the Examiner, Olson does not teach nor suggest the dealumination of a zeolite as presently claimed. It is applicants' position that Olson further in view of Eberly and Wakita does not teach or suggest a dealumination step to obtain the zeolite as presently claimed with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than 80 and less than 180, nor its use in an adsorption process.

Addressing the teachings of Eberly, the dealumination process taught by Eberly starts with a zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than about 3-5

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(Examples 1 through 6 of Eberly do not show a starting zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of more than 6) and converts to this starting zeolite to one with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio greater than about 5-10, preferably greater than about 20, and more preferably greater than about 50 (see Eberly, column 4, line 74 - column 5, line 5). The highest $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio achieved by Eberly was about 20 (see Example 5) and this was termed by Eberly as an "extremely high silica/alumina mole ratio of about 20". In the present invention, as can be seen from Examples 3 and 5, that the K-SSZ-13 was successfully dealuminated from a starting $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 20 to a final $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 160 while maintaining a zeolite of sufficient stability for separations. Obtaining a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 80 to 180 as presently claimed while obtaining a zeolite that has diffusion rate for a first component at least 50 times greater than the diffusion rate for a second component is simply not taught or suggested by Olson in view of Eberly.

Addressing the teachings of Wakita, the Examiner points to the teaching in Example 6 of the Wakita patent for teaching dealumination. Similar to Eberly, the starting point for the Wakita zeolite is an H-Y zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 5.5. There is no teaching by Wakita that $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios of 80 to 180 as presently claimed could be obtained by dealumination. In fact, Wakita does not even state what the final $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio is. One of ordinary skill in the art faced with the knowledge of Wakita and the teachings of Eberly would not be led to believe that the dealumination process in Wakita could achieve much more than the dealumination process of Eberly (i.e., final $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 20). Not only is this Si/Al molar ratio significantly lower than currently claimed, but the Examples in the current invention show that Si/Al ratios of about 20 simply do not work in the separation as claimed (see Example 3 and Figure 1 of the present application).

Therefore, it is applicants position that 1) in view of the teachings of Olson, one of skill in the art would not be motivated to attempt utilizing a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio less than about 200 in the separations process as presently claimed regardless of the teachings of Eberly and Wakita, and additionally, 2) that even in view of Wakita and Eberly, one of skill in the art would not be led to believe that

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obtaining a stable alkali metal containing zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios of 80 to 180 with a diffusion rate for a first component at least 50 times greater than the diffusion rate for a second component could be obtained via dealumination by the process as currently claimed.

Therefore, as expressed above, it is applicants' position that Claim 1 as presently amended with the limitations of prior Claim 4 is not obvious in light of Olson, Eberly, and Wakita. Claim 4 has been canceled and Claim 5 has been amended so that Claims 5 & 6 now depend from Claim 1. Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of present Claim 1 and all present dependent claims 2-3 and 5-9.

THIRD REJECTION UNDER 35 U.S.C. 103

The Examiner has rejected claims 1-3 and 7-9 under 35 U.S.C. 103(a) as being unpatentable over Ramachandran et al. (EP 0572239 A1) in view of Addiego (EP 0768111 A1).

Examiner's Position

It is the Examiner's position that Ramachandran discloses separating propylene from a mixture containing propane and propylene by using an adsorbent (e.g. 4A zeolite) having 8-membered tetrahedral rings. The adsorbent comprises alkali metal cations (e.g., sodium).

Ramachandran does not disclose a zeolite with silica to alumina molar ratio of greater than 50 but less than 200, does not disclose CHA or SSZ-13, and does not disclose that the cations are introduced by ion exchange at a pH greater than about 7.5.

It is Examiner's position that Addiego discloses an adsorbent with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of from about 50 to 250 and that it would have been obvious to one

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of ordinary skill in the art to have modified the Ramachandran process using the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio disclosed in Addiego.

The Examiner also states that it would have been obvious to have modified the Ramachandran process with a zeolite such as SSZ-13 because 4A zeolite has 8-member rings of tetrahedra. In addition, the Examiner is of the position that one of ordinary skill in the art would have modified the process of Ramachandran by introducing ion exchange as a pH greater than 7.5 because it is within the level of one of ordinary skill in the art to process the zeolite at the claimed pH.

Applicant's Position

It is applicants' position that the claims 1-3 and 7-9 as currently amended are not obvious in light of Ramachandran in view of Addiego.

In applicants' prior argument, it was argued that Ramachandran does not disclose nor teach a kinetic adsorption process as presently claimed. In Examiner's response, the Examiner is of the opinion that since the modified process of Ramachandran (by Addiego) would be essentially the same as the claimed process and it would be expected that some kinetic separation would occur.

Applicants respectfully again disagree, as there is no teaching that the Ramachandran process as modified would either result in a zeolite that has a diffusion rate for a first component at least 50 times greater than the diffusion rate for a second component (i.e., a highly kinetic separation). Although "some" kinetic separation may occur, there simply is no teaching that such a highly kinetic separation (such as the 50:1 ratio claimed) could be achieved. Addiego does not cure this deficiency as Addiego does not disclose or teach a kinetic process as currently claimed.

Additionally, it is applicants' position that Addiego actually teaches away from the kinetic separation as currently claimed. It is critical to the current process that the desired component be quickly adsorbed as well as quickly desorbed. Figure 2 of the current invention shows that the K-SSZ-13 with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 20

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quickly adsorbs propylene, but does not quickly desorb the propylene as required (i.e., after desorbing to about 60 mg/g of propylene, the desorption is very slow). Therefore, since the overall desorption process is slow, the K-SSZ-13 with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 20 does not possess sufficient desorption characteristics for a rapid cycle kinetic based process. The current invention solves this problem and it can be seen in Figure 3 that the K-SSZ-13 of the present invention with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 160 (i.e., within the claimed 80 to 180 molar ratio) has a very quick desorption down to about 20 mg/g of propylene and therefore is useful in a kinetic based separation process as claimed.

In contrast to the present invention, Addiego does not teach a rapid desorption kinetic based process, but teaches away from such a process. As can be seen in Addiego, "[t]he workstream is passed through the modified adsorber [*i.e.* modified with an acid-modifying agent] ... to cause adsorption and retention of the hydrocarbons. The adsorbed hydrocarbons can then be desorbed at a higher temperature than would otherwise occur with an unmodified adsorber." (see Addiego, Abstract, comments and emphasis added). It is the goal of Addiego to make the desorption of the adsorbed component slower and more difficult which is opposite of the objective of the current application. This contrary objective of Addiego invention is further exemplified in Addiego at page 2, line 57 - page 3, line 2 as well as page 5, lines 47-51 wherein it is clear that the objective of the Addiego alterations are to retard the desorption capabilities of the zeolite adsorbent.

Therefore, one of skill in the art would not look to Addiego which teaches modifications for reducing the desorption characteristics of a zeolite to arrive at the kinetic based process of the present invention which depends on increasing the desorption characteristics of the zeolite to operate in the kinetic based separations process as desired.

Secondly, Ramachandran utilizes a 4A zeolite in the process. The $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of a 4A is about 1.0. This is extremely lower than the zeolite $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios of about 80 to about 180 as presently claimed. Nothing in Ramachandran suggests the use or benefits of higher $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios nor does

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Ramachandran suggest dealuminating a zcolite to increase the $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio. It is applicants' position that that Addiego does not cure this deficiency as follows.

As Examiner points out, Addiego discloses the use of a zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 50 to 250. However, it is applicants' position that this is not the $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of the final zeolite utilized in the process of Addiego. Examiner is again reminded that the Olson patent taught away from utilizing zcolites with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio less than about 200 in the adsorption process. It is applicants' position that Addiego does not teach utilizing zeolites with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 80 to about 180 as presently claimed either. It is applicants' position that the Addiego reference also teaches away from a process utilizing a zeolite with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 80 to about 180 when the teaching of Addiego is properly viewed as a whole.

It is clear from reading Addiego that zeolites with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 50 to 250 must be modified (not utilized) to be useful in a separations process. This is explained on page 3, lines 18-22 where it is stated that "[t]he method of the present invention is to improve a hydrocarbon adsorber by modifying the acid sites or acidity of the adsorber. This is done by contacting the adsorber with an acid-modifying (or acid-site modifying) agent". It is later stated that that the zeolites well suited for the invention of Addiego are "zeolites having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of the zeolite is about 50 to 250" (Addiego, page 3, lines 42-43). This can be better understood by reviewing claims 1 through 5 of Addiego which state that the molecular sieve (i.e., zeolite) must be "modified" by contact with the acid-modifying agent prior to their use in a separations process. The full teaching of Addiego to one of skill in the art is contrary to the presently claimed invention and to the Examiner's use of the reference.

It is applicants' position that Addiego as with Olson, teaches away from the use of a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 80 to about 180. Addiego uses zeolites with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of about 50 to 250 as a starting point, and then modifies these zeolites so that they are useful in a separations process. As can be seen in Example 4 and Figure 1 of Addiego, the untreated zcolite does not adsorb/desorb

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propylene. Only after it is treated with an acid-modifying agent (i.e., phosphoric acid), does the zeolite possess sufficient adsorption/desorption properties. If the zeolites with a SiO₂/Al₂O₃ molar ratio of about 50 to 250 would be useful in a separations process, there would be no need for Addiego to modify them with an acid-modifying agent. Therefore, Addiego identifies the same problem that Olson is faced with and does not solve, i.e., that zeolites with the SiO₂/Al₂O₃ molar ratio of about 80 to about 180 as presently claimed are not useful in a separations process and therefore the prior art either discourages their use (as in Olson) or requires their modification prior to use (as in Addiego). What has presently been discovered is that through proper preparation, as presently disclosed and claimed, that an alkali metal containing zeolite with a SiO₂/Al₂O₃ molar ratio of about 80 to 180 can be utilized in a separations process and still maintain a high ratio of diffusivity (i.e., at least 50) between two components. This is not taught in the prior art and as explained above, and one of ordinary skill in the art with knowledge of the cited references would be dissuaded from attempting the separations process as presently claimed with a zeolite possessing the Si/Al molar ratios as presently claimed.

Applicants are also of the position that the Examiner has provided no evidence in either of the two responses to support his argument that one of ordinary skill in the art would have modified the Ramachandran process by introducing ion exchange at a pH of greater than about 7.5. Applicants therefore respectfully request that the Examiner provide support for the rejection of this claim or pass this claim to allowance.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of claims 1-3 and 7-9.

FOURTH REJECTION UNDER 35 U.S.C. 103

The Examiner has rejected claims 4-6 under 35 U.S.C. 103(a) as being unpatentable over Ramachandran et al. (EP 0572239 A1) and in view of Addiego (EP 0768111 A1) as in Claim 1 above and further in view of either Eberly et al. (3,591,488) or Wakita et al. (6,579,347).

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Examiner's Position

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It is the Examiner's position that Ramachandran and Addiego do not specifically disclose a step for dealuminating the zeolite by using steam. It is Examiner's position that both Eberly and Wakita teach a step of dealuminating a catalyst by steam and therefore it would have been obvious to one of ordinary skill in the art to have modified the combined processes of Ramachandran and Addiego by dealuminating the zeolite by steam as taught by either Eberly or Wakita.

Applicant's Position

As discussed in the "Third Rejection under 35 USC 103" above, Claim 1 has been amended to include the limitations of Claim 4 and Claim 4 has been canceled. For the reasons set out above, it is applicants' position that Claim 1 as currently amended is not obvious in light of the prior art and therefore Claims 5 & 6 as presently amended are patentable as claims depending from Claim 1.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejections of current claims 5-6.

In view of the foregoing remarks and the current amendments of the claims as presented, applicants respectfully request that the Examiner reconsider and withdraw the present claim objections and rejections and pass claims 1-3 and 5-9 as presently presented to allowance.

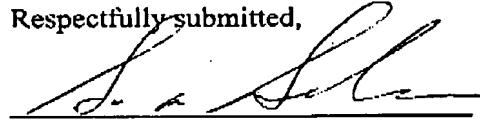
If the Examiner has any questions or requests information that would assist in progressing this application to allowance, the Examiner is encouraged to call applicants' attorney at phone number (908) 730-3210.

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If necessary to effect a timely response, this paper should be considered as a petition for an Extension of Time sufficient to effect a timely response. Please charge any deficiency in fees or credit any overpayments to **Deposit Account No. 05-1330.**

Respectfully submitted,



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Pursuant to 37 CFR 1.34(a)

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